

# PATENT COOPERATION TREATY

02 MAR 2005

From the  
INTERNATIONAL PRELIMINARY EXAMINING AUTHORITY

*Dr. Isenbruck*

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To:	
Dr. Günter Isenbruck Isenbruck, Bösl, Hörschler Wichmann, Huhn Theodor-Heuss-Anlage 12 68165 Mannheim ALLEMAGNE	Isenbruck   Bösl   Hörschler   Wichmann   Huhn. Patentanwälte Theodor-Heuss-Anlage 12 D-68165 Mannheim  - 3. Jan. 2005  Frist vortrist: 3.2.05 W.V.

NOTIFICATION OF TRANSMITTAL OF  
THE INTERNATIONAL PRELIMINARY  
EXAMINATION REPORT  
(PCT Rule 71.1)

Applicant's or agent's file reference B02/0522PC jw		<b>IMPORTANT NOTIFICATION</b>	
International application No. PCT/EP 03/10241	International filing date (day/month/year) 15.09.2003	Priority date (day/month/year) 16.09.2002	Date of mailing (day/month/year) 28.12.2004
Applicant BASF AKTIENGESELLSCHAFT et al.			

1. The applicant is hereby notified that this International Preliminary Examining Authority transmits herewith the international preliminary examination report and its annexes, if any, established on the international application.
2. A copy of the report and its annexes, if any, is being transmitted to the International Bureau for communication to all the elected Offices.
3. Where required by any of the elected Offices, the International Bureau will prepare an English translation of the report (but not of any annexes) and will transmit such translation to those Offices.


## 4. REMINDER

The applicant must enter the national phase before each elected Office by performing certain acts (filing translations and paying national fees) within 30 months from the priority date (or later in some Offices) (Article 39(1)) (see also the reminder sent by the International Bureau with Form PCT/IB/301).

Where a translation of the international application must be furnished to an elected Office, that translation must contain a translation of any annexes to the international preliminary examination report. It is the applicant's responsibility to prepare and furnish such translation directly to each elected Office concerned.

For further details on the applicable time limits and requirements of the elected Offices, see Volume II of the PCT Applicant's Guide.

The applicant's attention is drawn to Article 33(5), which provides that the criteria of novelty, inventive step and industrial applicability described in Article 33(2) to (4) merely serve the purposes of international preliminary examination and that "any Contracting State may apply additional or different criteria for the purposes of deciding whether, in that State, the claimed inventions is patentable or not" (see also Article 27(5)). Such additional criteria may relate, for example, to exemptions from patentability, requirements for enabling disclosure, clarity and support for the claims.

Name and mailing address of the international preliminary examining authority:   European Patent Office - P.B. 5818 Patentlaan 2 NL-2280 HV Rijswijk - Pays Bas Tel. +31 70 340 - 2040 Tx: 31 651 epo nl Fax: +31 70 340 - 3016	Authorized Officer  Delmon, G  Tel. +31 70 340-2525
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## PATENT COOPERATION TREATY

## PCT

## INTERNATIONAL PRELIMINARY EXAMINATION REPORT

(PCT Article 36 and Rule 70)



Applicant's or agent's file reference B02/0522PC jw	<b>FOR FURTHER ACTION</b> See Notification of Transmittal of International Preliminary Examination Report (Form PCT/IPEA/416)	
International application No. PCT/EP 03/10241	International filing date (day/month/year) 15.09.2003	Priority date (day/month/year) 16.09.2002
International Patent Classification (IPC) or both national classification and IPC B01J29/89		
Applicant BASF AKTIENGESELLSCHAFT et al.		

1. This international preliminary examination report has been prepared by this International Preliminary Examining Authority and is transmitted to the applicant according to Article 36.
2. This REPORT consists of a total of 6 sheets, including this cover sheet.  
  
☒ This report is also accompanied by ANNEXES, i.e. sheets of the description, claims and/or drawings which have been amended and are the basis for this report and/or sheets containing rectifications made before this Authority (see Rule 70.16 and Section 607 of the Administrative Instructions under the PCT).

These annexes consist of a total of 4 sheets.

3. This report contains indications relating to the following items:

- I ☒ Basis of the opinion
- II ☐ Priority
- III ☐ Non-establishment of opinion with regard to novelty, inventive step and industrial applicability
- IV ☐ Lack of unity of invention
- V ☒ Reasoned statement under Rule 66.2(a)(ii) with regard to novelty, inventive step or industrial applicability; citations and explanations supporting such statement
- VI ☐ Certain documents cited
- VII ☐ Certain defects in the international application
- VIII ☐ Certain observations on the international application

Date of submission of the demand  10.02.2004	Date of completion of this report  28.12.2004
Name and mailing address of the international preliminary examining authority:   European Patent Office - P.B. 5818 Patentlaan 2 NL-2280 HV Rijswijk - Pays Bas Tel. +31 70 340 - 2040 Tx: 31 651 epo nl Fax: +31 70 340 - 3016	Authorized Officer  Schoofs, B  Telephone No. +31 70 340-2760  

**INTERNATIONAL PRELIMINARY  
EXAMINATION REPORT**

International application No. PCT/EP 03/10241

**I. Basis of the report**

1. With regard to the **elements** of the international application (*Replacement sheets which have been furnished to the receiving Office in response to an invitation under Article 14 are referred to in this report as "originally filed" and are not annexed to this report since they do not contain amendments (Rules 70.16 and 70.17)*):

**Description, Pages**

1-26 as originally filed

**Claims, Numbers**

1-18 received on 18.05.2004 with letter of 17.05.2004

2. With regard to the **language**, all the elements marked above were available or furnished to this Authority in the language in which the international application was filed, unless otherwise indicated under this item.

These elements were available or furnished to this Authority in the following language: , which is:

- ☐ the language of a translation furnished for the purposes of the international search (under Rule 23.1(b)).
- ☐ the language of publication of the international application (under Rule 48.3(b)).
- ☐ the language of a translation furnished for the purposes of international preliminary examination (under Rule 55.2 and/or 55.3).

3. With regard to any **nucleotide and/or amino acid sequence** disclosed in the international application, the international preliminary examination was carried out on the basis of the sequence listing:

- ☐ contained in the international application in written form.
- ☐ filed together with the international application in computer readable form.
- ☐ furnished subsequently to this Authority in written form.
- ☐ furnished subsequently to this Authority in computer readable form.
- ☐ The statement that the subsequently furnished written sequence listing does not go beyond the disclosure in the international application as filed has been furnished.
- ☐ The statement that the information recorded in computer readable form is identical to the written sequence listing has been furnished.

4. The amendments have resulted in the cancellation of:

- ☐ the description, pages:
- ☐ the claims, Nos.:
- ☐ the drawings, sheets:

5. ☐ This report has been established as if (some of) the amendments had not been made, since they have been considered to go beyond the disclosure as filed (Rule 70.2(c)).

*(Any replacement sheet containing such amendments must be referred to under item 1 and annexed to this report.)*

6. Additional observations, if necessary:

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**INTERNATIONAL PRELIMINARY  
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**V. Reasoned statement under Article 35(2) with regard to novelty, inventive step or industrial applicability; citations and explanations supporting such statement**

**1. Statement**

Novelty (N)	Yes: Claims	
	No: Claims	1,16
Inventive step (IS)	Yes: Claims	
	No: Claims	2-15,17-18
Industrial applicability (IA)	Yes: Claims	1-18
	No: Claims	

**2. Citations and explanations**

**see separate sheet**

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**INTERNATIONAL PRELIMINARY  
EXAMINATION REPORT - SEPARATE SHEET**

International application No. PCT/EP 03/10241

**Re Item V**

**Reasoned statement with regard to novelty, inventive step or industrial applicability;  
citations and explanations supporting such statement**

1. Reference is made to the following document:

D1: WO 02 060580 A (TSUJI JUNPEI ;YAMAMOTO JUN (JP); SUMITOMO  
CHEMICAL CO (JP)) 8 August 2002 (2002-08-08) & EP 1 364 705 A  
(SUMITOMO CHEMICAL CO) 26 November 2003 (2003-11-26)

For the purpose of this communication, reference is made to EP 1 364 705 A.

2. The amended claims 1 to 18 as filed on 18.05.2004 are considered to be admissible in terms of Article 34(2)(b) PCT.

- 2.1 Independent claims 1 and 16 as amended now specify that  
(1) the solid material is intended "for use as catalytic material in an epoxidation reaction", and  
(2) "the at least one epoxide is the product of said epoxidation reaction".

3. From page 2, lines 18-23 of the description, it appears that the epoxide or hydrolysate thereof to be used in the present preparation process is 'the same' epoxide as the product of the epoxidation reaction wherein the solid material is to be used. In other words, if one intends to produce propylene oxide, one should use propylene oxide or propylene glycol in the preparation process. If however, one intends to produce ethylene oxide, one should use ethylene oxide or ethylene glycol in the preparation process.

- 3.1 However, such an interpretation is not in line with the wording of present claims 1 and 16 for the following reasons:

- 3.1(a) according to the wording of the claims, the solid material can be used in 'any' epoxidation reaction and hence the epoxide or hydrolysate thereof to be used in the preparation process can be the product of 'any' such epoxidation process.

**INTERNATIONAL PRELIMINARY  
EXAMINATION REPORT - SEPARATE SHEET**

International application No. PCT/EP 03/10241

As a result, the scope of the claim is not limited to the situation wherein the epoxide or hydrolysate thereof to be used in the preparation process is 'the same' epoxide as the product of the epoxidation reaction wherein the solid material is to be used.

- 3.1(b) If one were to interpret the expression "said epoxidation reaction" as used in claims 1 and 16 in a more restricting way such that 'the epoxide or hydrolysate thereof to be used in the preparation process is the product of the epoxidation reaction wherein the solid material is used', this would lead to a typical chicken and egg-situation since it would be impossible to prepare a catalyst for the first time when this preparation requires the use of a product that is imperatively obtained by using this - not yet existing - catalyst.
- 3.2 In view of the above, the wording of the claims is construed as indicated under paragraph 3.1(a) above.
4. D1 discloses a process for the preparation of a titanium-containing silicon oxide catalyst, in which diols such as ethylene glycol and the like are added as a solvent to the synthesis mixture containing the titanium source (D1, claim 1, example 1, paragraphs 15-26 and in particular paragraph 23).
- 4.1 D1 further discloses that the catalyst is separated from the mother liquor and molded into a shaped body. The catalyst is used in the epoxidation of olefins with hydroperoxides (D1, paragraphs 26-63).
- 4.2 Ethylene glycol is the hydrolysate of an epoxide which is the product of an epoxidation reaction.
- 4.3 The subject-matter of claim 1 is therefore not new (Article 33(2) PCT).
- 4.4 The further product by process features (Ia) to (If) in present claim 16 do not appear to be result in a feature in the resulting solid material that could distinguish it from the solid materials known from D1. The subject-matter of claim 16 is therefore not new (Article 33(2) PCT and PCT Guidelines, 5.26, 5.27 and A5.26).

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International application No. PCT/EP 03/10241

- 4.5 Dependent claims 2-15 and 17-18 do not appear to contain any features which, in combination with the features of any claim to which they refer, are associated with any technical effect that could support the presence of an inventive step. Hence, no inventive step is present in the subject-matter of claims 2-15 and 17-18 (Article 33(3) PCT).

Additional comment upon entry into the European phase

5. The expression "incorporated by reference" on page 22, lines 1-2 has to be deleted.
- 5.1 An inventive step has to be established over the whole claimed range, i.e. all epoxides or hydrolysates thereof, all transition metal oxides and all zeolites that come within the scope of the claims.

**We claim**

5

1. Process for preparing a solid material for use as catalytic material in an epoxidation reaction, the solid material containing at least one zeolite and being at least partially crystalline, wherein a step (I) of an at least partial crystallization of at least one solid material containing at least one zeolite out of a synthesis mixture involves at least one partial step of contacting at least one transition metal oxide source with at least one epoxide or hydrolysate thereof prior to or during the at least partial crystallization of said synthesis mixture into said solid material, and wherein the at least one epoxide is the product of said epoxidation reaction.

10

2. Process according to claim 1, characterised in that step (I) comprises at least the following partial steps

15

(Ia) mixing at least one hydrolyzable silicon source with a mineralizing and/or structuring agent and water;

20

(Ib) mixing at least one transition metal oxide source with an epoxide or a hydrolysate thereof;

25

(Ic) mixing the mixtures from (Ia) and (Ib) so that at least a part of the hydrolyzable compounds hydrolyzes;

30

(Id) distilling at least part of the alcohol that has been formed as a result of the at least partial hydrolysis of at least part of the hydrolyzable compounds;

(Ie) adding water to the bottom of (Id);

(If) reacting the synthesis mixture resulting from (Ie) at a temperature elevated with respect to room temperature.

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3. Process according to claim 2, characterized in that the hydrolyzable silicon source comprises at least one silicon oxide, the mineralizing and/or structuring agent comprises at least one tetraalkylammonium hydroxide, and the transition metal oxide source comprises at least one titanate.
4. Process according to claim 3, characterized in that the hydrolyzable silicon source comprises at least tetraethoxy silicate, the mineralizing and/or structuring agent comprises at least tetrapropylammonium hydroxide, the transition metal oxide source comprises at least tetrabutylorthotitanate and the epoxide or the hydrolysate thereof comprises at least propylene oxide or propylene glycol.
5. Process according to any one of claims 1 to 4, characterized in that the at least one zeolite belongs to at least one of the following structure classes: MFI, MEL, MWW, BEA or any mixed structure thereof.
6. Process according to any one of claims 1 to 5, step (I) of the at least partial crystallization resulting in a mixture (I) containing at least said solid material and a mother liquor, said process further comprising the step
- (II) separating and/or concentrating the solid material in mixture (I).
7. Process according to claim 6, characterized in that, after step (II), at least one of the following two additional steps is performed:
- (W) bringing the solid material from step (II) in contact with a composition containing water;
- (III) agglomerating or granulating or agglomerating and granulating the solid material from step (W) or from step (II).
8. Process according to claim 7, characterized in that, after step (W), the solid material is separated from at least part of the composition containing water.

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AMENDED SHEET

9. Process according to claim 6, optionally comprising the step

5 (III) agglomerating, or granulating, or agglomerating and granulating the solid material from step (II);

said process further comprising the step

10 (S) shaping the solid material from step (II) or (III) obtaining a shaped body.

10. Process according to claim 9, characterized in that the following step (W) is performed after step (II), or after step (S), or after step (II) and after step (S)

15 (W) bringing the solid material from step (II) or the shaped body from step (S) in contact with a composition containing water.

20 11. Process according to claim 9 or 10, characterized in that step (S) is selected from the group consisting of pelletizing, pressing, extruding, sintering, roasting, and briquetting.

12. Process according to claim 11, characterized in that before, or during, or before and during the step (S), a binding material is added to said solid material.

25 13. Process according to any one of claims 6 to 12, characterized in that after at least one of the steps (II), (W), (III) or (S), a step (C) of calcining the solid material, or the shaped body, or the solid material or the shaped body is performed.

30 14. Process according to claim 13, characterized in that step (C) is performed at temperatures higher than 400°C.

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AMENDED SHEET

15. Process according to any one of claims 6 to 14, characterized in that the process is an integrated process.

5 16. Solid material containing at least one zeolite for use as catalytic material in an epoxidation reaction, the solid material being obtainable by a process of treating a synthesis mixture, wherein a step (I) of an at least partial crystallization of at least one solid material containing at least one zeolite out of a synthesis mixture involves at least one partial step of contacting at least one transition metal oxide source with at least one epoxide or hydrolysate thereof prior to or during the at least partial crystallization of said synthesis mixture into said solid material, and wherein the at least one epoxide is the product of said epoxidation reaction, said step (I) comprising at least the following partial steps

15 (Ia) mixing at least one hydrolyzable silicon source with a mineralizing and/or structuring agent and water;

(Ib) mixing at least one transition metal oxide source with an epoxide or a hydrolysate thereof;

20 (Ic) mixing the mixtures from (Ia) and (Ib) so that at least a part of the hydrolyzable compounds hydrolyzes;

25 (Id) distilling at least part of the alcohol that has been formed as a result of the at least partial hydrolysis of at least part of the hydrolyzable compounds;

(Ie) adding water to the bottom of (Id);

30 (If) reacting the synthesis mixture resulting from (Ie) at a temperature elevated with respect to room temperature.

17. Solid material according to claim 16, characterized in that the solid material contains Ti.

35 18. Solid material according to claim 16 or 17, characterized in that the solid material is shaped into a shaped body.

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